Specific Heat of $Ce_{1-x}La_xRhIn_5$ in Zero and Applied Magnetic Field: A Very Rich Phase Diagram

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Abstract: Specific heat and magnetization results as a function of field on single- and poly-crystalline samples of $Ce_{1-x}La_xRhIn_5$ show 1.) a specific heat γ of about 100 mJ/moleK^2 (in agreement with recent dHvA results of Alvers et al.); 2.) upturns at low temperatures in C/T and χ that fit a power law behavior (<=> Griffiths phase non-Fermi liquid behavior); 3.) a field induced anomaly in C/T as well as M vs H behavior in good agreement with the recent Griffiths phase theory of Castro Neto and Jones, where M˜H at low field, M˜H$^\lambda$ above a crossover field, C/T˜T$^{-1+\lambda}$ at low field, and C/T˜(H$^{2+\lambda/2}$/T$^{3-\lambda/2}$)*exp(-μ_{eff}H/T) above the same crossover field as determined in the magnetization and where λ is independently determined from the temperature dependence of χ at low temperatures, χ˜T$^{-1+\lambda}$ and low fields.

I Introduction

Recently, a new family of heavy-fermion compounds has been discovered that crystallize in a layered, tetragonal structure with chemical composition $CeMIn_5$, where M= Ir, Co, and Rh. Characteristic of heavy-fermion systems, each member exhibits a large Sommerfeld coefficient γ ($\equiv$C/T as T$\rightarrow$0) in the specific heat C. $CeIrIn_5$ and $CeCoIn_5$ are bulk superconductors[1−2] with transition temperatures at T_c = 0.4 K and 2.3 K and normal-state values of $\gamma \approx 750$ mJ/molK^2 and 1200 mJ/molK^2, respectively. $CeRhIn_5$ displays heavy-fermion antiferromagnetism with[3] T_N = 3.8 K. A precise value of γ is difficult to establish unambiguously because of the Néel order; a lower limit of approximately 400 mJ/molK^2 has been quoted[4−5].

In our high field specific heat measurements[6] on the $CeMIn_5$ compounds, we found that the large upturn for M=Rh in C/T above T_N (C/T is already 1000 mJ/molK^2 at T_N) as temperature is lowered appeared to be primarily due to magnetic interactions above the antiferromagnetic transition since the specific heat data at a given temperature for T>T_N in different fields up to 32 T all coincide with one another when the temperature axis was scaled to T/T_N. Recently Alver, et al. have performed[7] dHvA measurements on twelve single crystal samples spanning the whole composition range of $Ce_{1-x}La_xRhIn_5$ and find rather low (i. e. inconsistent by approximately an order of magnitude with a γ of 400 mJ/molK^2) effective masses from the dilute Ce, large x end of the phase diagram up to x=0.1. At this Ce-rich end of the composition range they find an increase in the effective masses (which still remain $\leq$ 10 m_e) which they ascribe to spin fluctuation effects. Alver, et al. conclude that the Ce f-electrons remain localized in $Ce_{1-x}La_xRhIn_5$ for all x, with the (modest) observed mass enhancement near pure $CeRhIn_5$ due to spin fluctuation effects. Although comparisons between specific heat and dHvA data have inherent problems (not the least of which is the possibility of unseen, heavier mass orbits in the dHvA measurements), an effective mass enhancement of approximately ten normally corresponds to a specific heat γ of only ~50 mJ/molK^2. This is a wide discrepancy from the estimate of 400 mJ/molK^2 in[4−5] the literature; this discrepancy would be consistent with our high field specific heat result[6] that the upturn above T_N in C/T in pure $CeRhIn_5$ is primarily caused by magnetic interactions, which would not cause a mass enhancement observable, e.g., in dHvA measurements.

In order to help resolve this seeming disagreement, to determine the specific heat γ (also proportional to the effective mass) in a region of the phase diagram away from the antiferromagnetic anomaly, and to look for possible new behavior in the dilute limit we report here on a specific heat study of both single and polycrystalline samples of $Ce_{1-x}La_xRhIn_5$, 0$\leq$x$\leq$0.95. Certainly, doping studies[8−10] on other heavy Fermion systems, e.g. $Ce_{1-x}La_xCu_2Si_2$, $Ce_{1-x}Th_xCu_2Si_2$, and $U_{1-x}Th_xBe_{13}$, have revealed interesting new information - both about the respective parent compound as well as new physics in the dilute limit. Polycrystalline samples were originally chosen for the study as being more easily and rapidly prepared. However, specific heat results for polycrystalline

$Ce_{1-x}La_xRhIn_5$, x=0.5 and 0.8 were determined to disagree with specific heat results for single crystal samples, while results agreed for x=0.15 and 0.95. This disagreement appears due to the presence of a second phase which we were able to eliminate through long term annealing of the polycrystalline samples at a relatively low temperature.

II Experimental

Single crystal samples of $Ce_{1-x}La_xRhIn_5$ were prepared using the procedure described in ref. 6, which was similar to that used in refs. 4 and 7. Excess In was removed from the resulting flat platelet crystals using an H_2O:HF:H_2O_2 4:1:1 etch which was different than the centrifugal method (H_2O:HCl 4:1 etch) used in ref. 4 (7); however the present work's specific heat results (which are a measure of bulk properties) should be relatively independent of such surface treatments. The polycrystalline samples in the present work (previous work in the literature has been almost uniformly on single crystal samples) were prepared by melting together stoichiometric amounts of the appropriate high purity starting elements (using Ames Laboratory Ce and La, 99.95% pure Rh from Johnson Mathey Aesar, and 99.9999% In from Johnson Matthey Aesar - the same starting materials as used for the single crystals) under a purified inert Ar atmosphere. Weight losses after four melts, with a flipping of the arc-melted button between melts to improve homogeneity, were in the range of 1%, primarily due to In loss. Additional In was added in the beginning to correct for this, such that the In concentrations after the last melt were within ±0.2% of the stoichiometric amount.

Specific heat in fields to 13 T were measured using established techniques[11], while magnetic susceptibility data were measured in a SQUID magnetometer from Quantum Design.

III Results and Discussion

Figure 1 shows the specific heat divided by temperature vs temperature for single crystal $Ce_{1-x}La_xRhIn_5$, x=0, 0.15, 0.5, 0.8, and 0.95 and polycrystal $Ce_{1-x}La_xRhIn_5$, x=0.32. All samples were single phase. Results for unannealed polycrystalline $Ce_{1-x}La_xRhIn_5$, x=0.15 and 0.95, and annealed (35 days at 720 ^oC) polycrystalline $Ce_{1-x}La_xRhIn_5$, x=0.5 and 0.8, were comparable to the single crystal results (see inset of Fig. 1 for an example); however, unannealed polycrystalline samples for x=0.5 and 0.8 contained a second phase that ordered antiferromagnetically below 1 K. This was taken as a sign of an incipient miscibility gap which - due to previous work being focussed on single crystal samples - was heretofore unknown.

From the data shown in Fig. 1, one can follow the suppression of the antiferromagnetic transition with increasing La doping; there is a clear, although reduced in magnitude, transition at 2 K for 15% La doping that is absent by x=0.32. Although one might expect[12] non-Fermi liquid ('nFl') behavior when T_N is suppressed to T=0, the temperature dependence of the C/T data for

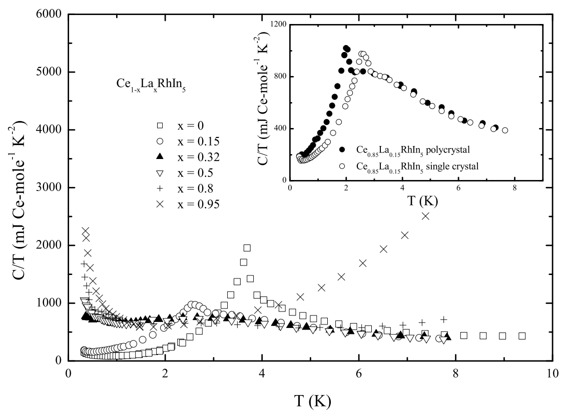

Figure 1: C/T vs T for Ce(1-x)La(x)RhIn(5)

x=0.32 - although the

data show an upturn - is only measured for ˜0.5 K below the hump. This is too restricted a temperature range to allow conclusions about the temperature dependence.

Before we discuss the behavior of γ as a function of x in $Ce_{1-x}La_xRhIn_5$, we will first focus on the upturn at low temperatures for x$\geq$0.5.

A Upturn in C/T for x$\geq$0.5

The upturn in C/T for x$\geq$0.5 in $Ce_{1-x}La_xRhIn_5$ shown in Figs. 1 is fit in Figs. 2 and 3 for single crystalline, as well as single phase polycrystalline, material. Note in Fig. 2 that the data for the three different samples agree rather well. There is certainly no sign in the dHvA results of Alver, et al. for a strong, heavy fermion upturn in C/T that would cause large effective masses. Thus, this upturn at low temperatures in C/T likely has a magnetic interaction explanation (see section **C** below for the field dependence). The temperature dependence of the upturns in C/T (see Figs. 2 and 3) for single crystal $Ce_{1-x}La_xRhIn_5$, x=0.5, 0.8, and 0.95, is not at all like the high temperature side of a Schottky peak (C ˜1/T^2) but rather appears (in the somewhat limited temperature range that we have data) to follow C/T ˜$T^{-1+\lambda}$,$\lambda_{C/T}$=0.63 $\pm$ 0.1, 0.37 $\pm$ 0.1, and ˜0 respectively. This is the temperature dependence predicted for non-Fermi liquid behavior caused by disorder-induced spin clusters, the so-called Griffiths phase[12−13]. (Note that the fits of χ to $T^{-1+\lambda}$ below 1.2 K are much better than fits to either log T or $T^{0.5}$.) In this theory, the magnetic susceptibility

at low temperature should have the same power law dependence as C/T. The susceptibility at low temperatures for these same compositions of single

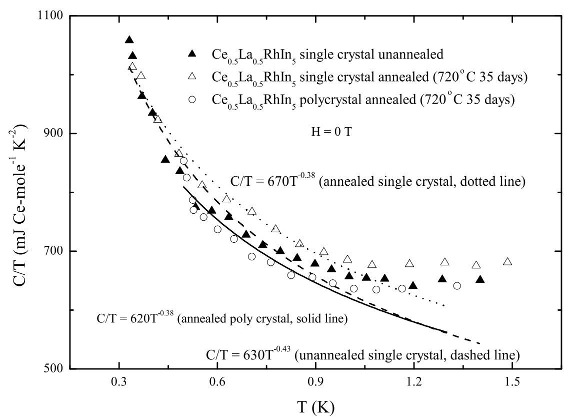

Figure 2: C/T for 3 samples of x=0.5

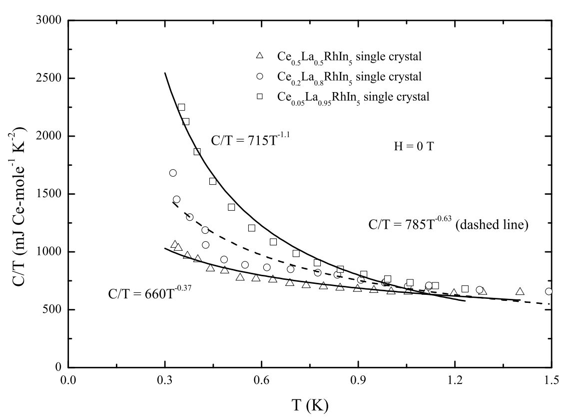

Figure 3: C/T vs T, x=0.5,0.8,0.95, fit to Tˆ(-1+lambda)

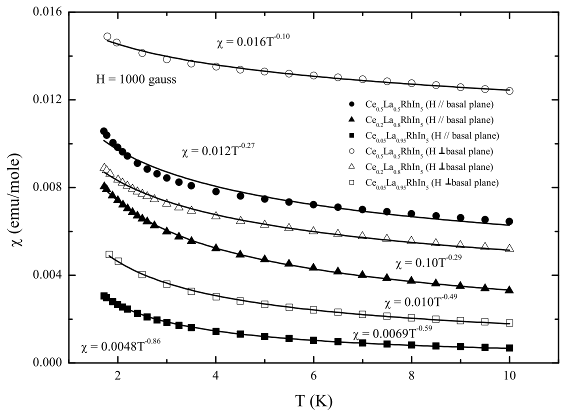

Figure 4: Chi vs T fit to T^(-1+lambda)

crystal $Ce_{1-x}La_xRhIn_5$, see Fig. 4, does indeed fit this $T^{-1+\lambda}$ temperature dependence, with λ_χ ={0.73,0.90}, {0.50,0.70}, {0.14,0.30} respectively for H {⊥,‖} the c-axis, where the absolute error bar for each value is ± 0.1 (with, however, somewhat better precision, useful for intercomparison between values derived from a *given* measurement technique. For example, 0.14 derived from χ for x=0.95 is certainly less than 0.30 derived for the other field direction, but is comparable to the value of ˜0 derived for the same composition from the specific heat.) (Note that other standard non-Fermi liquid temperature dependences, such as χ ˜log T or $T^{0.5}$, do not fit the χ data at all well.) Although for a given composition the respective exponents for C/T and χ agree within experiment accuracy only for χ(H ⊥ c), the recent theory[14] of Castro Neto and Jones actually predicts that χ and C/T may diverge *differently* at low temperature, relaxing the requirement of the early theory[12−13] that $\lambda_\chi = \lambda_{C/T}$. It is clear that the disorder requirement for uncompensated spins (which requires that M vs H is shows saturation behavior) is fulfilled for all these compositions (see discussion and accompanying figures in section **C** below.) In addition, the agreement in $\lambda_{C/T}$ and $\lambda_\chi$found for the upturn in C/T and χ in the present work is comparable to that found by, e. g., deAndrade et al.[15] in their study of $Th_{1-x}U_xPd_2Al_3$ - even though they measured χ down to 0.5 K, i. e. in a temperature range comparable to that for their specific heat measurements. The anisotropy of the susceptibility-determined λ values is thought to be real, and not related to the discrepancy between $\lambda_{C/T}$ and λ_χ.

As one possible check for a tendency towards magnetic behavior, the Wilson ratio (R $\propto \chi/\gamma\mu_{eff}^2$) - which is used[16] in the study of heavy Fermion systems to track the tendency towards magnetism, with R $\gtrsim$ 0.8 indicating[16] magnetic behavior - for these $Ce_{1-x}La_xRhIn_5$ alloys is in the range of 1.0 to 1.8, i. e. they definitely show magnetic character. As a further check for evidence

for spin clusters, we investigated these compositions for spin glass behavior and - to within the limits (±2 %) of the accuracy of the measurements - found no difference between field cooled and zero field cooled data down to 1.8 K. This lack of observable spin glass behavior in the dc magnetic susceptibility in these samples does not rule out a Griffiths phase interpretation[17].

B Specific Heat γ as a Function of x

The original goal of this work, besides the hope for new physics of interest in the dilute range (already partially fulfilled by the results discussed above for the low temperature upturn in C/T and χ) was to investigate the specific heat γ (defined as C/T as T$\rightarrow$0) away from the region of the phase diagram where antiferromagnetism obscures C/T as T$\longrightarrow$0 in $CeRhIn_5$ diluted with La. As discussed above, after the antiferromagnetism is suppressed (x>0.15), a low temperature upturn in the C/T data (Fig. 1) occurs that, normalized per Ce-mole, becomes more pronounced with increasing dilution of the Ce. This upturn appears not to be related to the effective masses measured by the dHvA measurements.

A further complication to determining the specific heat γ is the rounded feature in C/T centered at ˜3 K visible already for x=0.15 above T_N. As may be seen from Fig. 5, the C/T data for x=0.5 (triangles) and 0.8 (circles) in $Ce_{1-x}La_xRhIn_5$ above the low temperature upturn show a tendency to curve or bend downwards down to about 1.5 K, at which point the upturn discussed in the section above begins. This 'hump' in C/T centered at ˜3 K makes extrapolating C/T to T=0 to determine γ a somewhat imprecise procedure. It should be stressed that this rounded feature, or hump, in C/T has its provenance in the f-electron sublattice: such a feature is *not* present in C/T data for pure $LaRhIn_5$[18]. One possibility for correcting for this feature in order to determine γ is to subtract off both the low temperature upturn (see Fig. 3 for the fits to the upturns) *and* a fit[18] to pure $LaRhIn_5$ and examine the remainder. As shown in the inset to Fig. 5 for x=0.5, this very rough approximation (the apparent negative value below about 1 K is, see Fig. 3, merely a sign that the fit to the upturn - which goes up to over 1000 mJ/Ce-moleK^2 at 0.3 K - is in error as T$\rightarrow$1 K) allows us to assign an approximate[19] γ value per Ce mole of $\leq$100 mJ/CemolK^2 for x$\geq$0.5. This agrees much better with Alver, et al.'s dHvA results than the estimates of 400 mJ/CemolK^2 estimated[4−5] in the literature. However, as the La dilution is removed, for x$\leq$0.1, Alver, et al. report approximately a factor of two increase in effective mass due to spin fluctuation effects, with an effective mass for pure $CeRhIn_5$ that would correspond to a γ of approximately 50 mJ/CemolK^2. In the dilute limit, Alver et al.'s effective measured effective mass corresponds to a γ of only 25 mJ/CemolK^2. However, as may be seen in Fig. 5, our C/T data at low temperature are much too obscured by the unexpected upturn as well as by the rounded maximum to supply any sort of accurate estimate for γ beyond the dilute, x$\geq$0.5, range of $\leq$100 mJ/CemolK^2 already quoted above.

C Field Induced Anomaly for x$\geq$0.5

As a final aspect of new, unexpected behavior for $CeRhIn_5$ diluted with

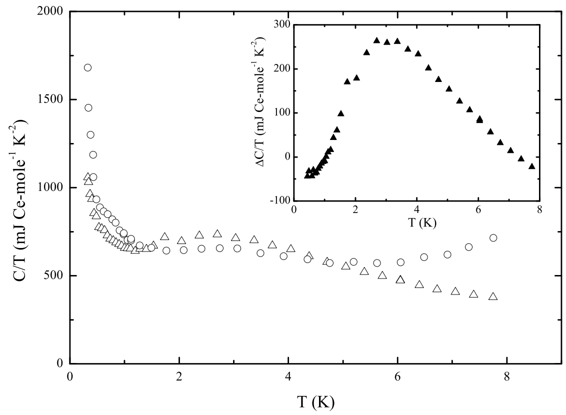

Figure 5: C/T vs T showing the 'hump' at 3 K

La, when we were investigating the field dependence of the upturn in the specific heat divided by temperature using magnetic field as a probe, we discovered that applied field suppresses the low temperature upturn in C/T at rather low field and induces an peak in C/T that, with increasing field, moves up in temperature and becomes broader and less pronounced. This rounded anomaly, shown in Fig. 6 for x=0.95 (these data are typical of the results for all x$\geq$0.5) with field in the basal plane (data in the perpendicular direction are within 15 percent of these), is not that of either a spin glass (where C˜1/T above the peak) or a Schottky anomaly (C˜1/T^2 above the peak) but rather seems to be a field-induced anomaly. (The upturns in C/T for H $\geq$ 6 T are caused by the applied field splitting the nuclear magnetic moment energy levels and creating a Schottky peak in the specific heat.)

Castro Neto and Jones have recently published[14] a theory of how the specific heat and magnetization of materials with non-Fermi liquid behavior caused by disorder-induced Griffiths phase spin clusters should scale with magnetic field. In general, both the magnetization and specific heat are predicted to exhibit low field behaviors (M ˜H and C/T ˜T$^{-1+\lambda}$) which crossover over to the respective high field behaviors (M ˜H$^\lambda$ and C/T ˜(H$^{2+\lambda/2}$/T$^{3-\lambda/2}$)e$^{-\mu_{eff}H/T}$) at the *same* magnetic field. The prediction for the field and temperature dependence for the high field specific heat leads to a peak in C/T (or a shoulder in C) as a function of increasing temperature - thus qualitatively consistent with the data shown in Fig. 6.

Although the specific heat data in field was taken in fairly widely spaced fields, the fact that a peak occurs already in C/T in H=3 T offers a prediction (the equality of the crossover field requires that the crossover field for the magnetization data be perforce below 3 T) that can be checked by examining the M vs H data, where a much more finely spaced sequence of fields was used. In

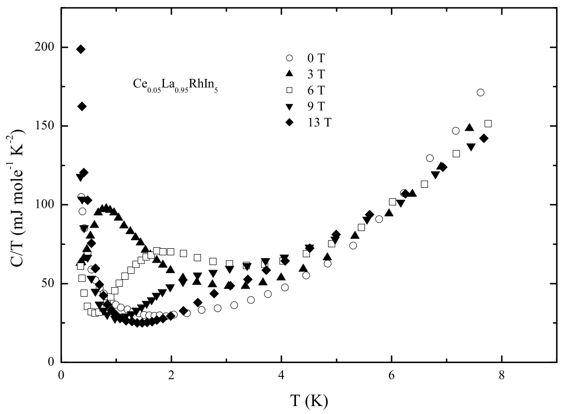

Figure 6: Field-induced anomaly in C/T for x=0.95

addition, the high field prediction that M ˜H^{λ} can be checked up to 5.5 T, and this field-dependence determination of λ can then be compared with that independently determined from the *temperature dependence* of χ in Figure 4. Thus, magnetization data for both field directions for single crystal $Ce_{0.05}La_{0.95}RhIn_5$ are shown fitted to these Griffiths phase low and high field predictions in Figures 7 and 8, H $\|,\perp$ basal plane respectively. As may be seen, using the values for λ_χ determined from Fig. 4 (0.14 and 0.41 for H($\|,\perp$) basal plane respectively) gives rather good[20] agreement between the predicted, M ˜H^{λ} dependence and the high field magnetization data. (The fit to the higher field data with the lowest standard deviation actually gives λ=0.67; however, the standard deviations are within 8% of one another.) Further, the deviation from linear behavior at low fields occurs (see Figs. 7 and 8) above 0.8 T and the deviation from the M ˜H^{λ} power law occurs below 1.2 T. These estimates for the crossover field are not inconsistent with the peak in C/T (where a peak is characteristic of the high field regime) occuring in 3 T, Fig. 6. (Work under way[21] to more thoroughly characterize the low and high field behavior for M and C/T for x=0.95 has found that a peak in C/T field data taken in 0.5 T increments down to 0.3 K first appears at 1.5 T.)

Another prediction[14] of the Griffiths phase theory of Castro Neto and Jones, the field and temperature dependence of C/T in the high field limit, is compared[22] to the 3T $Ce_{0.05}La_{0.95}RhIn_5$ data (with the fit[18] to pure $LaRhIn_5$ and the small, <10% at the lowest temperature, contribution due to the field splitting of the nuclear moments, subtracted off), H $\|$ basal plane, in Fig. 9. Using only two fit parameters (the amplitude and the effective moment, μ_{eff}) and fixing $\lambda = 0.14$ (based on λ_χ) gives the fit (dashed line in Fig. 9) as shown, with the reasonable[14,23] fitted value for μ_{eff} (which corresponds to the average

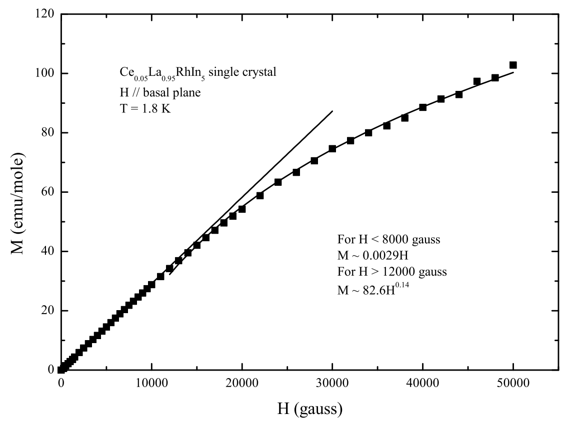

Figure 7: M vs H for x=0.95, H in basal plane

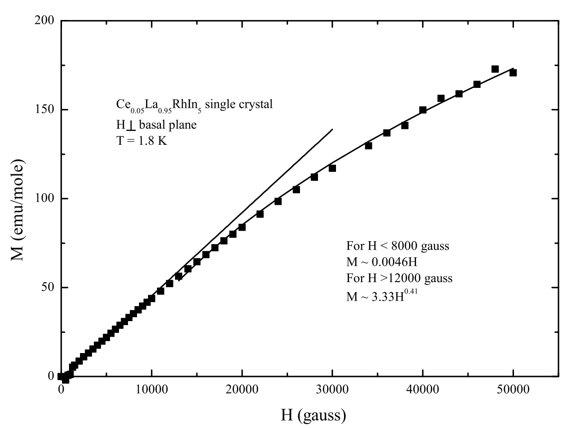

Figure 8: M vs H for x=0.95, H perp. to basal plane

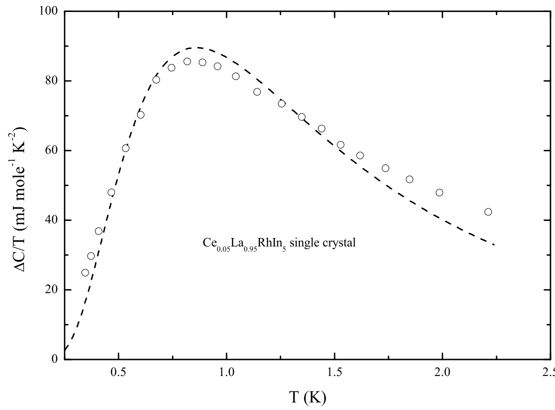

Figure 9: Fit of field-induced anomaly to theory

moment in the Griffiths phase spin cluster) of 1.25 μ_B. Clearly, fitting C/T to $(H^{2+\lambda/2}/T^{3-\lambda/2})e^{-\mu_{eff}H/T}$ is a fairly good representation of the data. (To give an idea how the fit depends on the effective moment, a fit to these 3 T data with μ_{eff} constrained to be 1.0 μ_B is shifted by to lower temperatures by ˜0.2 K from the present fit.)

IV Conclusions

Despite the difficulty of precisely compensating for the broad peak in C/T in $Ce_{1-x}La_xRhIn_5$ centered at about 3 K, the apparent γ per Ce mole for x≥0.5, away from the antiferromagnetic transition in the phase diagram, appears to be less than 100 mJ/Ce-moleK^2 - in disagreement with estimates for γ in the literature[4−5] but not inconsistent with the dHvA results of Alvers et al.[7] There is a strong upturn in C/T below 1 K for x≥0.5 that, when compared to the temperature dependence of the susceptibility and the non-linear M vs H data, is consistent with non-Fermi liquid behavior due to disordered spin clusters ('Griffiths phases.') Applied magnetic field suppresses this upturn in C/T already by 3 T; above 3 T the C/T results show a broad anomaly that further broadens and moves to higher temperatures as field is increased. This field induced anomaly, together with the field dependence of the magnetization, compares well with the predictions of the Griffiths phase theory[14,24] of Castro Neto and Jones, particularly in the magnetization data as a function of field and the agreement of these data with the predicted λ_χ exponent from the temperature dependence of the susceptibility. In summary, the breadth of behavior observed in $Ce_{1-x}La_xRhIn_5$ in zero and applied field is indicative of a phase diagram of unusual richness and variety.

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17. For a discussion of spin cluster, spin glass, and Griffiths phase behavior, see ref. 12.

18. Mike Hundley, private communication. See also ref. 4.

19. Note of course the unavoidable uncertainty is fitting the 'hump' - which may very well involve entropy due to Ce-Ce interactions - to data from a more dilute composition and then applying this fit to more concentrated systems.

20. The "best fit" value for the exponent λ from the field dependence of the magnetization for H $\parallel$ basal plane shown in Fig. 7 is within 0.1 of the value $\lambda_\chi = 0.14$ determined from the temperature dependence of χ determined in Fig. 4, i. e. within the error bar. For H $\perp$ basal plane, the best fit to the magnetization data shown in Fig. 8 gives $\lambda = 0.67$ instead of the value determined from the temperature dependence of χ, where $\lambda_\chi = 0.41$. However, the standard deviation for the fit (to 20 data points) using $\lambda_\chi = 0.41$ is less than 8% higher than that for the "best" fit.

21. J. S. Kim, J. Alwood, D. Mixson, and G. R. Stewart, to be published.

22. Fits to the 6 and 9 T data are similar, although the correction for the low temperature upturn in C/T caused by the nuclear hyperfine level splitting due to the applied field is larger and the size of the field-induced anomaly in C/T with increasing field is rapidly decreasing. Since the crossover field between low and high field dependences, as determined by the magnetization, is ˜ 0.8 -1.2 T, the 3 T data should be well in the high field limit.

23. A. H. Castro Neto, private communication.

24. Although a recent paper (A. J. Millis, D. K. Morr, and J. Schmalian, Phys. Rev. Lett. **87**, 167202 {2001}) has called the theory of Castro Neto and Jones into question based on dissipation arguments in the single impurity limit, an even more recent work by Castro Neto and Jones (cond-mat/0106176) argues that for concentrated systems the results of ref. 14 still hold.